

Wang and Brady Reply: We thank Ikeda, Berthier, and Sollich (IBS) [1] for their comments which question our interpretation of the universal viscosity divergence near the flow-arrest transition in constant stress and pressure rheology of hard-sphere colloidal suspensions [2]. IBS introduced two Péclet numbers: $Pe_0 = \dot{\gamma}a^2/d_0$ and $Pe = \dot{\gamma}a^2/d(\phi)$, with $\dot{\gamma}$ the strain rate, a the particle size, d_0 the isolated single-particle diffusivity, and $d(\phi)$ the long-time at-rest self-diffusivity. And they considered three regimes: (i) $Pe_0 < Pe \ll 1$, (ii) $Pe_0 \ll 1 \ll Pe$, and (iii) $1 \ll Pe_0 < Pe$.

IBS's claim that "only Pe is considered in [2]" is not true. The stress Péclet number $Pe_\sigma = \sigma a^2/(\eta_0 d_0)$, with σ the imposed stress and η_0 the solvent viscosity, is a primitive input to our simulations. It compares the magnitude of the imposed stress relative to the particle thermal fluctuations and is trivially connected to Pe_0 through $Pe_\sigma = \eta Pe_0$, with η the dimensionless shear viscosity.

Near the flow-arrest transition, Pe_0 is of little relevance to suspension dynamics. What drive an otherwise arrested suspension to flow are internal structural rearrangements, which are characterized by $d(\phi)$, not by the local "in cage" thermal fluctuations described by d_0 . Near athermal jamming, i.e., close to the point (ϕ_{SAP}, μ_{SAP}) in Fig. 1, the condition $Pe_0 \gg 1$ is not satisfied. Here, the imposed pressure $\bar{\Pi} = Pe_\sigma/(6\pi\mu_{SAP})$ satisfies $\bar{\Pi} \sim (\phi_{SAP} - \phi)^{-\delta}$ with $\delta = 1$ near jamming [3]. Meanwhile, the universal viscosity divergence suggests $Pe_0 \sim Pe_\sigma(\phi_{SAP} - \phi)^\gamma$ with $\gamma \approx 2$, which leads to $Pe_0 \sim \mu_{SAP}(\phi_{SAP} - \phi)^{\gamma-\delta}$, independent of Pe_σ and $\bar{\Pi}$. Thus, $Pe_0 \ll 1$ for $\gamma > \delta$, which is the case for hard-sphere suspensions when $(Pe_\sigma, \phi) \rightarrow (\infty, \phi_{SAP})$. IBS's distinction between regimes (ii) and (iii) is therefore unnecessary, and Pe alone is sufficient. This is also reflected in recent experiments [4] which show that suspensions enter the non-Brownian regime sooner, i.e., at

lower Pe_0 , with increasing ϕ : The shear stresses where the shear thinning regime ends are the same over a wide range of ϕ .

In regime (i), linear response theory requires $\Pi(\phi, \dot{\gamma}) = \Pi^{eq}(\phi) + \Delta\Pi(\phi)\dot{\gamma}^2$ and $\sigma(\phi, \dot{\gamma}) = \eta_T(\phi)\dot{\gamma}$. Because of the different $\dot{\gamma}$ dependences, one can always evaluate $\eta_T(\phi)$ at sufficiently small $\dot{\gamma}$ with $\Pi \approx \Pi^{eq}(\phi)$. In the low μ limit, constant Π and constant ϕ results are equivalent. This is shown in Fig. 1: Far from the glass transition ϕ_g , the contour at constant Π_1 asymptotes to the contour at constant ϕ_1 at a low but finite μ . Near ϕ_g , the contours at Π_2 and ϕ_2 approach each other as $\mu \rightarrow 0$. Therefore, by construction, our approach can probe the glass transition. On the other hand, the viscosity divergences observed along constant- ϕ and constant- Π contours may be different due to the different approaches to the arrested region [2], as illustrated by the viscosity contours in Fig. 1. Furthermore, it is still an open question whether the product $\eta_T(\phi)d(\phi)$ remains constant near ϕ_g , and, consequently, simulations and experiments of the relaxation time [5] cannot infer the viscosity divergence [1].

When the at-rest volume fraction is above ϕ_g , the diffusivity $d(\phi) \rightarrow 0$ and the suspension has a yield stress. This corresponds to IBS's regimes (ii) and (iii). Here, the viscosity is inherently non-Newtonian regardless of Pe_0 and exhibits universal divergences at constant Π . IBS's interpretation using a Herschel-Bulkey model for the pressure nicely complements our work. Our study is for true hard spheres whose behavior can be fundamentally different from soft-particle systems, even when the stiffness of the potential is increased [6] or the confining pressure is reduced [7] to eliminate particle overlaps. For example, in the non-Brownian limit, the singular hard-sphere potential leads to a finite shear viscosity despite the stress's thermal origin [8]. In the same limit, the viscosity from a soft potential (no matter how stiff) approaches zero.

Finally, we agree with IBS that in their regime (iii), our data are sparse since ϕ_{SAP} can only be approached from below in our simulations. However, as we have already pointed out, $Pe_0 \gg 1$ cannot be achieved near athermal jamming, and our results agree with the viscosity divergence found in non-Brownian experiments [9].

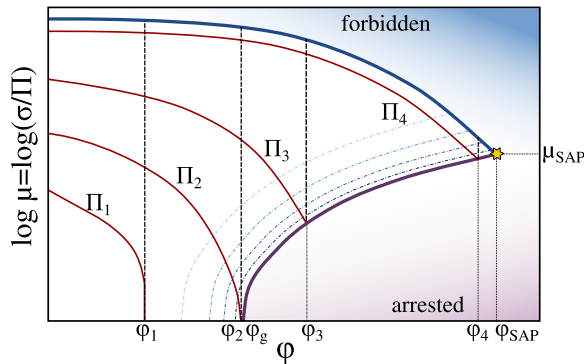


FIG. 1. Sketch of the μ - ϕ flow map based on Ref. [2]. The thick curves enclose the flowing region, with the lower curve outlining the arrested region and the upper curve outlining the non-Brownian limiting behavior. The two curves intersect at the shear arrest point (μ_{SAP}, ϕ_{SAP}) . The solid lines represent constant- Π contours at pressures $\Pi_1 < \Pi_2 < \Pi_3 < \Pi_4$. The dashed lines show the constant- ϕ contours at the corresponding at-rest volume fraction. The dash-dotted lines are the constant-viscosity contours.

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